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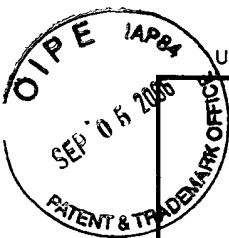


PTO/SB/21 (05-03)

Approved for use through 04/30/2003. OMB 0651-0031

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TRANSMITTAL FORM

(to be used for all correspondence after initial filing)

Issued Patent No.	7,074,308
Issue Date	July 11, 2006
Application Number	10/714,835
Filing Date	November 14, 2003
First Named Inventor	MAO, FEI
Group Art Unit	1753
Examiner Name	OLSEN, KAJ K
Attorney Docket Number	ADCI-001CON

Total Number of Pages in This Submission 27

ENCLOSURES (check all that apply)

<input type="checkbox"/> Fee Transmittal Form	<input type="checkbox"/> Assignment Papers (for an Application)	<input type="checkbox"/> After Allowance Communication to Group
<input type="checkbox"/> Fee Attached	<input type="checkbox"/> Drawing(s)	<input type="checkbox"/> Appeal Communication to Board of Appeals and Interferences
<input type="checkbox"/> Amendment / Reply	<input type="checkbox"/> Licensing-related Papers	<input type="checkbox"/> Appeal Communication to Group (Appeal Notice, Brief, Reply Brief)
<input type="checkbox"/> After Final	<input type="checkbox"/> Petition	<input checked="" type="checkbox"/> Petition for Certificate of Correction (3 pgs.)
<input type="checkbox"/> Affidavits/declaration(s)	<input type="checkbox"/> Petition to Convert to a Provisional Application	<input checked="" type="checkbox"/> Certificate of Correction (2 pgs.)
<input type="checkbox"/> Extension of Time Request	<input type="checkbox"/> Power of Attorney, Revocation Change of Correspondence Address	<input checked="" type="checkbox"/> Other Enclosure(s) (please identify below):
<input type="checkbox"/> Express Abandonment Request	<input type="checkbox"/> Terminal Disclaimer	Pages 3 & 7 of the Application as filed on 11/14/03 showing correction
<input type="checkbox"/> Information Disclosure Statement	<input type="checkbox"/> Request for Refund	Copy of Response filed on 6/15/05 (16 pgs.)
<input type="checkbox"/> Certified Copy of Priority Documents	<input type="checkbox"/> CD, Number of CD(s)	Copies of last pages of issued patent with changes (3 pgs.)
<input type="checkbox"/> Response to Missing Parts/ Incomplete Application		
<input type="checkbox"/> Response to Missing Parts under 37 CFR 1.52 or 1.53		

Remarks

SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT

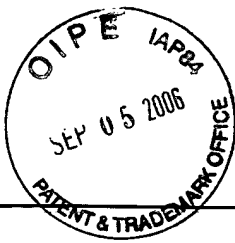
Signing Attorney/Agent (Reg. No.)	EDWARD J. BABA, 52,581 BOZICEVIC, FIELD & FRANCIS, LLP	Certificate
Signature		SEP 12 2006
Date	September 5, 2006	of Correction

EXPRESS MAIL LABEL NO. EV 686 489 752 US

This collection of information is required by 37 CFR 1.5. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

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SEP 13 2006



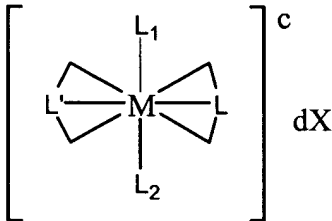
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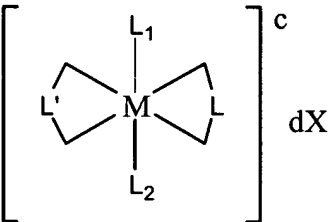
<p align="center">PETITION FOR CERTIFICATE OF CORRECTION UNDER 37 C.F.R. § 1.322 FOR PATENT AND TRADEMARK OFFICE ERROR</p> <p>Address to: Mail Stop DAC Commissioner for Patents P.O. Box 1450 Alexandria, VA 22313-1450</p>	Attorney Docket Number	ADCI-001CON
	First Named Inventor	FEI MAO
	Application Number	10/714,835
	Filing Date	November 14, 2003
	Patent Number	7,074,308
	Issue Date	July 11, 2006
	Title	TRANSITION METAL COMPLEXES WITH (PYRIDYL)IMIDAZOLE LIGANDS

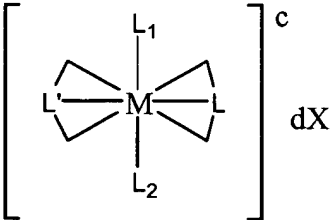
Sir:

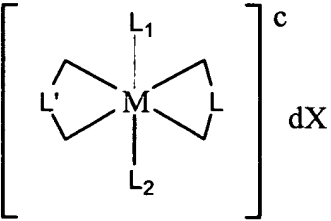
Applicants petition under 37 C.F.R. § 1.322 for a Certificate of Correction to correct errors in the specification and claims for the above-identified patent due to Patent and Trademark Office error.

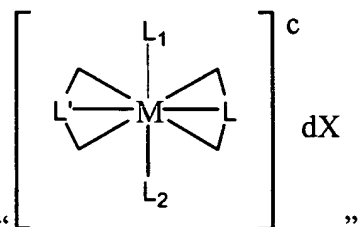
Transmitted herewith for filing is a Certificate of Correction for the above-identified patent. Please make the following corrections to Claims 1 and 15, and the specification at columns 2 and 5.

In column 2, lines 37-45, please replace the formula “” with the

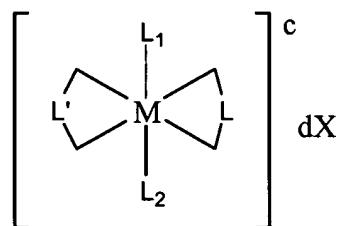
formula --  --.

In column 5, lines 7-15, please replace the formula “” with the

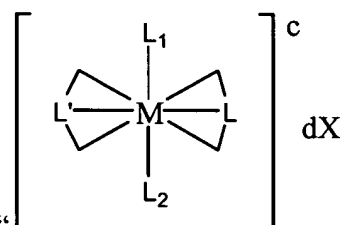
formula --  --.



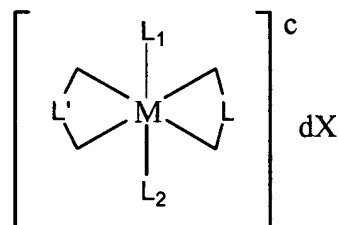
In claim 1, column 37, lines 3-11, please replace the formula “



with the formula --



In claim 15, column 38, lines 9-17, please replace the formula “




with the formula --

Enclosed are copies of pages 3 and 7 of the patent application as filed on November 14, 2003 showing the correct formula, and a copy of the Response to the Office Action filed on June 15, 2005, showing the correct formula in renumbered claims 1 and 15 (originally claims 49 and 63). Also enclosed, is a copy of the last pages of the issued patent showing the incorrect language of the specification and the claims that resulted from Patent and Trademark Office error.

It is believed that no fee is due since the error was made by the Patent and Trademark Office. However, the Commissioner is hereby authorized to charge any fees under 37 C.F.R. § 1.20 which may be required by this paper, or to credit any overpayment, to Deposit Account No. 50-0815.

Respectfully submitted,
BOZICEVIC, FIELD & FRANCIS LLP

Date: Sept. 5, 2004

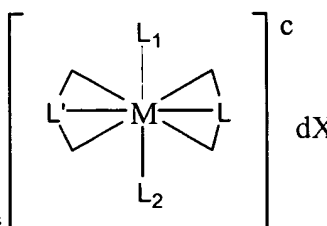
By: 
Edward J. Baba
Registration No. 52,581

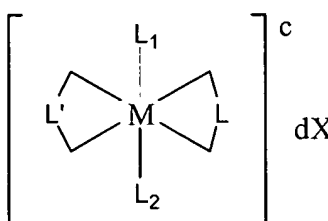
BOZICEVIC, FIELD & FRANCIS LLP
1900 University Avenue, Suite 200
East Palo Alto, CA 94303
Telephone: (650) 327-3400
Fax: (650) 327-3231

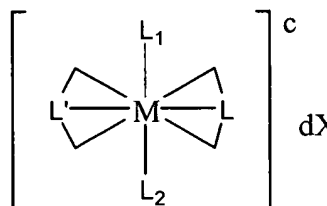
UNITED STATES PATENT AND TRADEMARK OFFICE CERTIFICATE OF CORRECTION

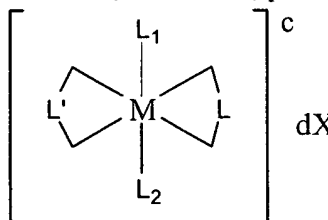
PATENT NO. : 7,074,308
 DATED : July 11, 2006
 INVENTOR(S) : Fei Mao, Adam Heller

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

In column 2, lines 37-45, please replace the formula "" with the

formula --  --.

In column 5, lines 7-15, please replace the formula "" with the

formula --  --.

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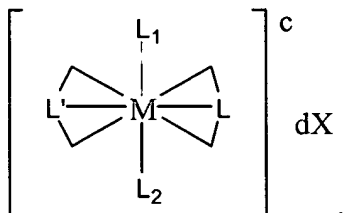
PATENT NO: 7,074,308

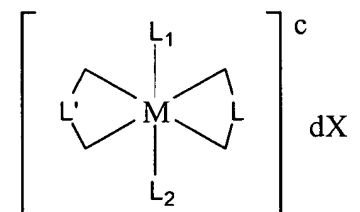
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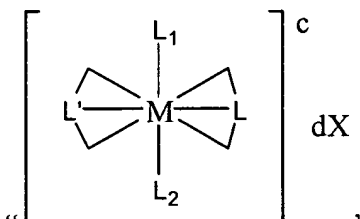
UNITED STATES PATENT AND TRADEMARK OFFICE CERTIFICATE OF CORRECTION

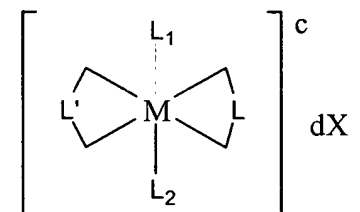
PATENT NO. : 7,074,308
 DATED : July 11, 2006
 INVENTOR(S) : Fei Mao, Adam Heller

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

In claim 1, column 37, lines 3-11, please replace the formula “”

with the formula ----.

In claim 15, column 38, lines 9-17, please replace the formula “”

with the formula ----.

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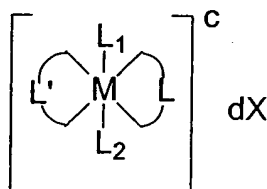
PATENT NO: 7,074,308

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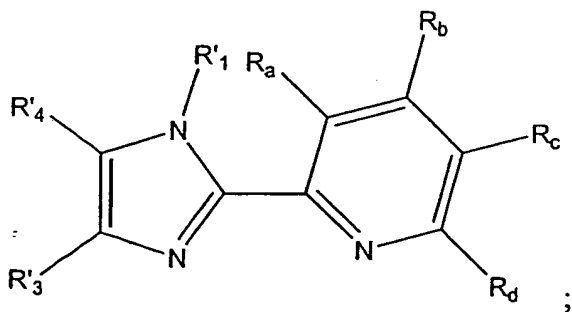
Summary of the Invention

[0007] The present invention is directed to novel transition metal complexes. The present invention is also directed to the use of the complexes as redox mediators. The preferred redox mediators typically exchange electrons rapidly with enzymes and electrodes, are stable, can be readily synthesized, and have a redox potential that is tailored for the electrooxidation of analytes, such as glucose for example.

[0008] One embodiment of the invention is a transition metal complex having the general formula set forth below.



In this general formula, M is cobalt, iron, ruthenium, osmium, or vanadium; c is an integer selected from -1 to -5, 0, or +1 to +5 indicating a positive, neutral, or negative charge; X represents at least one counter ion; d is an integer from 0 to 5 representing the number of counter ions, X; L and L' are independently selected from the group consisting of:

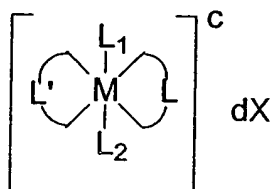


and L₁ and L₂ are other ligands. In the formula for L and L', R₁ is a substituted or an unsubstituted alkyl, alkenyl, or aryl group. Generally, R₃, R₄, R_a, R_b, R_c, and R_d are independently -H, -F, -Cl, -Br, -I, -NO₂, -CN, -CO₂H, -SO₃H, -NHNH₂, -SH, -OH, -NH₂, or substituted or unsubstituted alkoxycarbonyl, alkylaminocarbonyl, dialkylaminocarbonyl, alkoxy, alkylamino, dialkylamino, alkanoylamino, arylcarboxamido, hydrazino, alkylhydrazino, hydroxylamino, alkoxyamino, alkylthio, alkenyl, aryl, or alkyl.

[0009] The transition metal complexes of the present invention are effectively employed as redox mediators in electrochemical sensors, given their very fast kinetics. More particularly,

effective reagents in synthesis than other transition metal redox mediators.

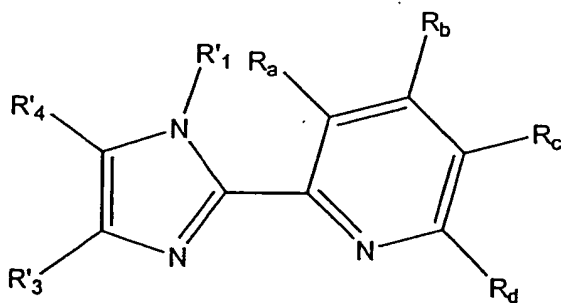
[0026] Compounds having Formula 1, set forth below, are examples of transition metal complexes of the present invention.



1

[0027] M is a transition metal and is typically iron, cobalt, ruthenium, osmium, or vanadium. Ruthenium and osmium are particularly suitable for redox mediators.

[0028] L and L' are each bidentate, substituted or unsubstituted 2-(2-pyridyl)imidazole ligands having the Structure 2 set forth below.



2

[0029] In Structure 2, R'1 is a substituted or an unsubstituted aryl, alkenyl, or alkyl. Generally, R'1 is a substituted or an unsubstituted C1-C12 alkyl or alkenyl, or an aryl, such as



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Applicants: Mao et al.
Title: Transition of Metal Complexes with (Pyridyl) Imidazole Ligands
Application No.: 10/714,835
Filing Date: November 14, 2003
Conf. No.: 3222
Atty Docket No.: THER.001US1

ENCLOSED:

1. This Return Receipt Postcard
2. Transmittal Letter (1 page - in duplicate)
3. Amendment (9 pages)
4. Terminal Disclaimer to Obviate a Double Patenting Rejection Over a Patent (3 pages)
5. Statement Under 37 CFR 3.73(b) (1 page)
6. Check for \$130.00

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JUN 15 2005

Applicants: Mao et al.
Title: Transition of Metal Complexes with (Pyridyl) Imidazole Ligands
Application No.: 10/714,835
Filing Date: November 14, 2003
Conf. No.: 3222
Atty Docket No.: THER.001US1

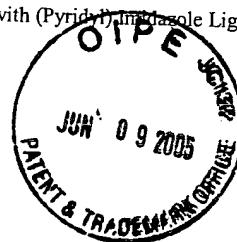
Parsons Hsue & de Huntz LLP

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6. Check for \$130.00

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PARSONS HSUE & DE RUNTZ
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(415) 318-1160

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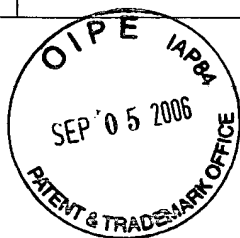
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THER.001US1	\$130.00
*2000-001	\$130.00

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THER.001US1	PATENT: Terminal Disclaimer filing fee



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June 7, 2005

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Commissioner For Patents
P.O. Box 1450

Alexandria, VA 22313-1450

Re: Applicant(s): Mao et al.
 Title: Transition Metal Complexes with (Pyridyl) Imidazole Ligands
 Application No.: 10/714,835 Filing Date: November 14, 2003
 Examiner: Kaj K. Olsen Group Art Unit: 1753
 Docket No.: THER.001US1 Conf. No.: 3222

Dear Sir:

Transmitted herewith are the following documents in the above-identified application:

- (1) Return Receipt Postcard;
- (2) This Transmittal Letter (1 page - in duplicate);
- (3) Amendment (9 pages);
- (4) Terminal Disclaimer to Obviate a Double Patenting Rejection Over a Patent (3 pages);
- (5) Statement Under 37 CFR 3.73(b) (1 page); and
- (6) Check for \$130.00.



No additional fee is required.



The fee has been calculated as shown below:

CLAIMS AS AMENDED

	Claims Remaining <u>After Amendment</u>		Highest No. Previously <u>Paid For</u>		Present <u>Extra</u>		<u>Rate</u>		Additional <u>Fee</u>
Total Claims	25	Minus	70	=	0	x	\$50.00	\$	0.00
Independent Claims	2	Minus	3	=	0	x	\$200.00	\$	0.00
<input type="checkbox"/>	Fee of \$360.00 for the first filing of one or more multiple dependent claims per application							\$	
<input checked="" type="checkbox"/>	Fee for Terminal Disclaimer to Obviate a Double Patenting Rejection Over a Patent							\$	130.00
Total additional fee for this Amendment:								\$	130.00



Conditional Petition for Extension of Time: If an extension of time is required for timely
filing of the enclosed document(s) after all papers filed with this transmittal have been
considered, an extension of time is hereby requested.



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Signature

Respectfully submitted,

K. Alison de Runtz
Reg. No. 37,119



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s): Mao et al.
Title: Transition Metal Complexes with (Pyridyl) Imidazole Ligands
Serial No.: 10/714,835 Filed: November 14, 2003
Examiner: Kaj K. Olsen Group Art Unit: 1753
Docket No.: THER.001US1 Conf. No.: 3222

Certificate of Mailing Under 37 CFR 1.8

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Signature Gulen Bower

Mail Stop Amendment
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

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AMENDMENT

Sir:

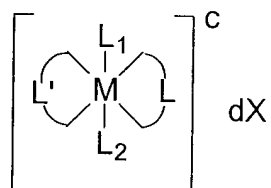
This is in response to an office action that was mailed on May 12, 2005 and set a shortened statutory period for response that expires on August 12, 2005. In view of the following amendments and remarks, reconsideration is respectfully requested.

In the Claims:

Please amend Claims 65, 67, 69 and 71, such that the claims are as set forth below.

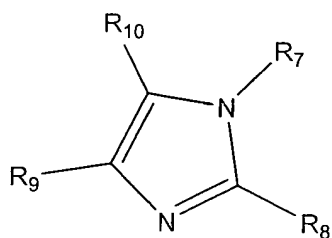
Claims 1-48. (Cancelled)

49. (Previously Presented) A complex having the formula:



wherein M is osmium;

L_1 has the formula:



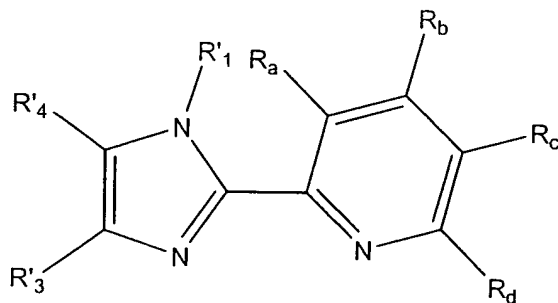
wherein R_7 is a substituted or unsubstituted aryl or a substituted or unsubstituted C1-C12 alkyl;

R_8 is $-H$ or a C1 alkyl; and

a combination of R_9 and R_{10} forms a fused, saturated or unsaturated, 5- or 6-membered ring;

L_2 is a halide;

L and L' are independently:



wherein R'_1 is a substituted or unsubstituted C1-C6 alkyl;

R'_3 and R'_4 are independently $-H$; and

R_a , R_b , R_c , and R_d are independently $-H$ or $C1$ alkyl;
 c is a $+1$ or $+2$ charge;
 X is an anion; and
 d is a number of X sufficient to balance the charge c .

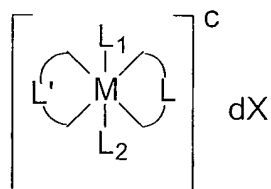
50. (Previously Presented) The complex of claim 49, wherein R_7 is a $C5$ alkyl.
51. (Previously Presented) The complex of any one of claims 49 and 50, wherein R_8 is methyl.
52. (Previously Presented) The complex of any one of claims 49 and 50, wherein a combination of R_9 and R_{10} forms a fused, saturated or unsaturated, 6-membered ring.
53. (Previously Presented) The complex of any one of claims 49 and 50, wherein a combination of R_9 and R_{10} forms a fused, unsaturated, 6-membered ring.
54. (Previously Presented) The complex of any one of claims 49 and 50, wherein L_2 is $-F$, $-Cl$, or $-Br$.
55. (Previously Presented) The complex of any one of claims 49 and 50, wherein L_2 is $-Cl$.
56. (Previously Presented) The complex of any one of claims 49 and 50, wherein R'_1 is a $C1$ - $C2$ alkyl.
57. (Previously Presented) The complex of any one of claims 49 and 50, wherein R'_1 is a $C1$ alkyl.
58. (Previously Presented) The complex of any one of claims 49 and 50, wherein each of R_a and R_c is $-H$.
59. (Previously Presented) The complex of any one of claims 49 and 50, wherein each of R_a , R_b , and R_c is $-H$.

60. (Previously Presented) The complex of any one of claims 49 and 50, wherein each of R_a , R_b , R_c , and R_d is $-H$.

61. (Previously Presented) The complex of any one of claims 49 and 50, wherein X is a halide.

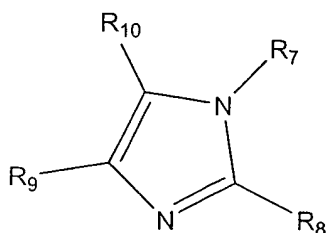
62. (Previously Presented) The complex of any one of claims 49 and 50, wherein X is chloride.

63. (Previously Presented) A complex having the formula:



wherein M is osmium;

L_1 has the formula:



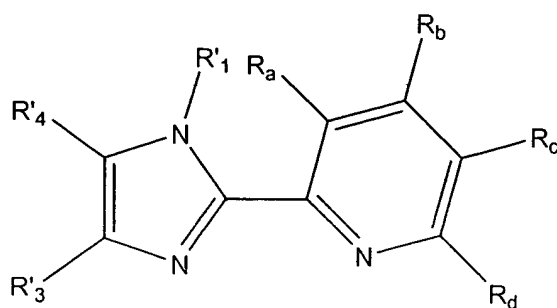
wherein R_7 is a C1-C12 alkyl;

R_8 is $-H$; and

a combination of R_9 and R_{10} forms a fused, unsaturated, 6-membered ring;

L_2 is chloride;

L and L' are independently:



wherein R'₁ is methyl; and

R'₃, R'₄, R_a, R_b, R_c, and R_d are independently -H;

c is +2;

d is 2; and

X is chloride.

64. (Previously Presented) The complex of claim 63, wherein R₇ is a C5 alkyl.

65. (Currently Amended) The complex of any one of claims 49, 50, 63, and 64, wherein at least one of ligand selected from L, L', L₁ and L₂ is coupled to a polymeric backbone.

66. (Previously Presented) The complex of claim 65, wherein the polymeric backbone comprises at least one functional group that is a ligand of the complex.

67. (Currently Amended) The complex of claim 66, wherein the functional group is selected from ~~a group consisting of~~ pyridine and imidazole groups.

68. (Previously Presented) A sensor comprising:
 a working electrode;
 a counter electrode;
 an enzyme disposed proximate to the working electrode; and
 the complex of any one of claims 49, 50, 63, and 64 disposed proximate to the working electrode.

69. (Currently Amended) The sensor of claim 68, wherein the complex is coupled to a polymeric backbone via at least one of ligand selected from L, L', L₁ and L₂.

70. (Previously Presented) The sensor of claim 69, wherein the polymeric backbone comprises at least one functional group that is a ligand of the complex.

71. (Currently Amended) The sensor of claim 70, wherein the functional group is selected from ~~a group consisting of~~ pyridine and imidazole groups.

72. (Previously Presented) The sensor of claim 68, wherein the complex is crosslinked on the working electrode.

73. (Previously Presented) The sensor of claim 68, wherein the complex and the enzyme are crosslinked on the working electrode.

In the Specification:

Please delete paragraph [0001] as previously amended and replace it with the replacement paragraph set forth below, which is marked to show changes being made.

This application is a continuation of and claims priority based on United States Patent Application No. 10/143,300 of Mao *et al.*, filed on May 9, 2002 and entitled "Transition Metal Complexes with (Pyridyl)imidazole Ligands and Sensors Using Said Complexes," which issued as United States Patent No. 6,676,816 on January 13, 2004 and is related to United States Provisional Patent Application No. 60/290,537 of Fei Mao, filed on May 11, 2001 and entitled "Transition Metal Complexes with (Pyridyl)imidazole Ligands[[],]." Each of the foregoing applications each of which is incorporated herein in its entirety by this reference.

REMARKS

Each of Claims 65, 67, 69 and 71 has been amended merely to remove unnecessary language therein, and in some cases, to replace same with other language. Each of Claims 65, 67, 69 and 71 is no narrower by virtue of its amendment herein. No new matter has been added by virtue of these amendments.

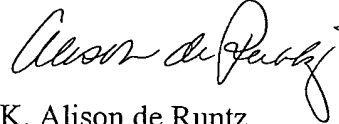
The specification has been amended to reflect the maturity of United States Patent Application No. 10/143,300 into United States Patent No. 6,676,816. No new matter has been added by virtue of this amendment to the specification.

Claims 49-73 have been rejected under the judicially created doctrine of obviousness-type double patenting over Claims 1-3, 6-9, 11-13, 16, 17, 25-31, 34-36, 39, 40, 44, 46-49, 61 and 66-70 of United States Patent No. 6,676,816. A terminal disclaimer concerning same is submitted herewith. This submission of the terminal disclaimer simply serves an expedient as to the removal of these rejections. It is not an admission or an acquiescence, and raises neither a presumption nor an estoppel, as to the merits of these rejections. *Quad Environmental Technologies Corp. v. Union Sanitary District*, 946 F.2d 870, 20 USPQ2d 1392 (Fed.Cir. 1991). It is believed that these rejections have been overcome.

The Office's indication that upon resolution of the above-mentioned obviousness-type double patenting rejections, Claims 49-73 are allowable, is acknowledged and much appreciated. The Office has provided a statement of reasons for the indication of allowable subject matter. As to United States Patent No. 6,338,790 to Feldman *et al.* (hereinafter, "Feldman"), it is submitted that same does not preclude patentability of Claims 49-73 pursuant to 35 U.S.C. Section 103(c)(1). As such, no comment of the Office concerning Feldman, other than the reference to common ownership, is addressed or conceded herein. As to the comments of the Office as to United States Patent Nos. 4,421,751 and 4,382,872, and the articles of Yu *et al.* and Gholamkhass *et al.*, it is submitted that the teaching and/or lack thereof as to each of these references is as contained in each of these references. It is submitted that Claims 49-73 are allowable in view of the art of record on the basis that there is no teaching or suggestion in same of the invention of each of Claims 49-73.

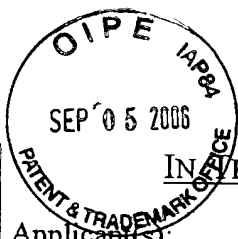
In view of the foregoing, an early indication that the application is in condition for allowance is earnestly solicited.

Respectfully submitted,

A handwritten signature in cursive script, appearing to read "Alison de Runtz".

K. Alison de Runtz
Reg. No. 37,119

PARSONS HSUE & DE RUNTZ LLP
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San Francisco, CA 94111
(415) 318-1160
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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s): Fei Mao and Adam Heller
Title: Transition Metal Complexes with (Pyridyl) Imidazole Ligands
Serial No.: 10/714,835 Filing Date: November 14, 2003
Examiner: Kaj K. Olsen Group Art Unit: 1753
Docket No.: THER.001US1 Conf. No.: 3222

Certificate of Mailing Under 37 CFR 1.8

I hereby certify that this correspondence is being deposited with the United States Postal Service with sufficient postage as first class mail in an envelope address to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on 6/7/05

Signature Julien Bowe

Commissioner of Patents
P.O. Box 1450
Alexandria, VA 22313-1450

TERMINAL DISCLAIMER TO OBVIATE A
DOUBLE PATENTING REJECTION OVER A PATENT

Sir:

Assignee, TheraSense, Inc., a Corporation having a place of business at 1360 South Loop Road, Alameda, CA 94502, is the owner of the entire interest in the instant application. Assignee hereby disclaims, except as provided below, the terminal part of the statutory term of any patent granted on the instant application, which would extend beyond the expiration date of the full statutory term defined in 35 U.S.C. §§ 154 to 156 and 173, as presently shortened by any terminal disclaimer, of prior U.S. Patent No. 6,676,816. Assignee hereby agrees that any patent so granted on the instant application shall be enforceable only for and during such period that it and the prior patent are commonly owned. This agreement runs with any patent granted on the instant application and is binding upon the grantee, its successors or assigns.

In making the above disclaimer, Assignee does not disclaim the terminal part of any patent granted on the instant application that would extend to the expiration date of the full statutory term as defined in 35 U.S.C. §§ 154 to 156 and 173 of the prior patent, as presently shortened by any terminal disclaimer, in the event that it later: expires for failure to pay a maintenance fee, is held unenforceable, is found invalid by a court of competent jurisdiction, is statutorily disclaimed in whole or terminally disclaimed under 37 C.F.R. § 1.321, has all claims canceled by a reexamination certificate, is reissued, or is in any manner terminated prior to the expiration of its full statutory term as presently shortened by any terminal disclaimer.

The undersigned (whose title is supplied below) represents that he/she is a representative authorized to sign on behalf of Assignee. A Certificate for Taking Action accompanies this document.

The undersigned declares that all statements made herein of my own knowledge are true, all statements made herein on information and belief are believed to be true, and all statements made herein are made with the knowledge that whoever, in any matter within the jurisdiction of the Patent and Trademark Office, knowingly and willfully falsifies, conceals, or covers up by any trick, scheme, or device a material fact, or makes any false, fictitious or fraudulent statements or representations, or makes or uses any false writing or document knowing the same to contain any false, fictitious or fraudulent statement or entry, shall be subject to the penalties including fine or imprisonment or both as set forth under 18 U.S.C.

1001, and that violations of this paragraph may jeopardize the validity of the application or this document, or the validity or enforceability of any patent, trademark registration, or certificate resulting therefrom.

5/31/05
Date

CSJ
Signature

Carl Silverman
Typed Or Printed Name

Divisional Vice President Intellectual Property
Title



Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

STATEMENT UNDER 37 CFR 3.73(b)

Applicant/Patent Owner: TheraSense, Inc.

Application No./Patent No.: 6,676,816 Filed/Issue Date: January 13, 2004

Entitled: Method for Removing Photoresist from Low-K Films in a Downstream Plasma System

TheraSense, Inc., a Corporation
(Name of Assignee) (Type of Assignee, e.g., corporation, partnership, university, government agency, etc.)

states that it is:

1. ☒ the assignee of the entire right, title, and interest; or
2. ☐ an assignee of less than the entire right, title and interest.
The extent (by percentage) of its ownership interest is _____ %

in the patent application/patent identified above by virtue of either:

A. ☒ An assignment from the inventor(s) of the patent application/patent identified above. The assignment was recorded in the United States Patent and Trademark Office at Reel 013083, Frame 0242, or for which a copy thereof is attached.

OR

B. ☐ A chain of title from the inventor(s), of the patent application/patent identified above, to the current assignee as shown below:

1. From: _____ To: _____
The document was recorded in the United States Patent and Trademark Office at
Reel _____, Frame _____, or for which a copy thereof is attached.
2. From: _____ To: _____
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Reel _____, Frame _____, or for which a copy thereof is attached.
3. From: _____ To: _____
The document was recorded in the United States Patent and Trademark Office at
Reel _____, Frame _____, or for which a copy thereof is attached.

☐ Additional documents in the chain of title are listed on a supplemental sheet.

☐ Copies of assignments or other documents in the chain of title are attached.

[NOTE: A separate copy (i.e., a true copy of the original assignment document(s)) must be submitted to Assignment Division in accordance with 37 CFR Part 3, if the assignment is to be recorded in the records of the USPTO. See MPEP 302.08]

The undersigned (whose title is supplied below) is authorized to act on behalf of the assignee.

Carl Silverman
Signature

Carl Silverman
Printed or Typed Name

Divisional Vice President Intellectual Property
Title

5/31/05
Date
(510) 549-5479
Telephone Number

This collection of information is required by 37 CFR 3.73(b). The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.11 and 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

If you need assistance in completing the form, call 1-800-PTO-9199 and select option 2.

TRANSITION METAL COMPLEXES WITH (PYRIDYL)IMIDAZOLE LIGANDS

CROSS-REFERENCE TO RELATED APPLICATIONS

This application is a continuation of and claims priority based on U.S. patent application Ser. No. 10/143,300 of Mao, et al., filed on May 9, 2002 and entitled "Transition Metal Complexes with (Pyridyl)imidazole Ligands and Sensors Using Said Complexes," which issued as U.S. Pat. No. 6,676,816 on Jan. 13, 2004 and is related to U.S. Provisional Patent Application No. 60/290,537 of Fei Mao, filed on May 11, 2001 and entitled "Transition Metal Complexes with (Pyridyl)imidazole Ligands." Each of the foregoing applications is incorporated herein in its entirety by this reference.

FIELD OF THE INVENTION

This invention relates to transition metal complexes with (pyridyl)imidazole ligands. In addition, the invention relates to the preparation of the transition metal complexes and to the use of the transition metal complexes as redox mediators.

BACKGROUND OF THE INVENTION

Enzyme-based electrochemical sensors are widely used in the detection of analytes in clinical, environmental, agricultural and biotechnological applications. Analytes that can be measured in clinical assays of fluids of the human body include, for example, glucose, lactate, cholesterol, bilirubin and amino acids. Levels of these analytes in biological fluids, such as blood, are important for the diagnosis and the monitoring of diseases.

Electrochemical assays are typically performed in cells with two or three electrodes, including at least one measuring or working electrode and one reference electrode. In three electrode systems, the third electrode is a counter-electrode. In two electrode systems, the reference electrode also serves as the counter-electrode. The electrodes are connected through a circuit, such as a potentiostat. The measuring or working electrode is a non-corroding carbon or metal conductor. Upon passage of a current through the working electrode, a redox enzyme is electrooxidized or electroreduced. The enzyme is specific to the analyte to be detected, or to a product of the analyte. The turnover rate of the enzyme is typically related (preferably, but not necessarily, linearly) to the concentration of the analyte itself, or to its product, in the test solution.

The electrooxidation or electroreduction of the enzyme is often facilitated by the presence of a redox mediator in the solution or on the electrode. The redox mediator assists in the electrical communication between the working electrode and the enzyme. The redox mediator can be dissolved in the fluid to be analyzed, which is in electrolytic contact with the electrodes, or can be applied within a coating on the working electrode in electrolytic contact with the analyzed solution. The coating is preferably not soluble in water, though it may swell in water. Useful devices can be made, for example, by coating an electrode with a film that includes a redox mediator and an enzyme where the enzyme is catalytically specific to the desired analyte, or its product. In contrast to a coated redox mediator, a diffusional redox mediator, which can be soluble or insoluble in water, functions by shuttling electrons between, for example, the enzyme and the electrode. In any case, when the substrate of the enzyme is

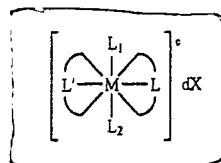
electrooxidized, the redox mediator transports electrons from the substrate-reduced enzyme to the electrode; and when the substrate is electroreduced, the redox mediator transports electrons from the electrode to the substrate-oxidized enzyme.

Recent enzyme-based electrochemical sensors have employed a number of different redox mediators such as monomeric ferrocenes, quinoid compounds including quinines (e.g., benzoquinones), nickel cyclamates, and ruthenium amines. For the most part, these redox mediators have one or more of the following limitations: the solubility of the redox mediators in the test solutions is low, their chemical, light, thermal, and/or pH stability is poor, or they do not exchange electrons rapidly enough with the enzyme or the electrode or both. Some mediators with advantageous properties are difficult to synthesize. Additionally, the redox potentials of some of these reported redox mediators are so oxidizing that at the potential at which the reduced mediator is electrooxidized on the electrode, solution components other than the analyte are also electrooxidized. Some other of these reported redox mediators are so reducing that solution components, such as, for example, dissolved oxygen, are also rapidly electroreduced. As a result, the sensor utilizing the mediator is not sufficiently specific.

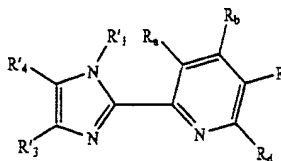
SUMMARY OF THE INVENTION

The present invention is directed to novel transition metal complexes. The present invention is also directed to the use of the complexes as redox mediators. The preferred redox mediators typically exchange electrons rapidly with enzymes and electrodes, are stable, can be readily synthesized, and have a redox potential that is tailored for the electrooxidation of analytes, such as glucose for example.

One embodiment of the invention is a transition metal complex having the general formula set forth below.



In this general formula, M is cobalt, iron, ruthenium, osmium, or vanadium; c is an integer selected from -1 to -5, 0, or +1 to +5 indicating a positive, neutral, or negative charge; X represents at least one counter ion; d is an integer from 0 to 5 representing the number of counter ions; L and L' are independently selected from the group consisting of:

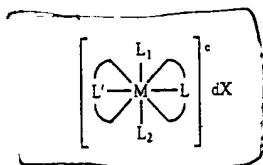


and L₁ and L₂ are other ligands. In the formula for L and L', R₁ is a substituted or an unsubstituted alkyl, alkenyl, or aryl group. Generally, R₃, R₄, R₅, R₆, R₇, and R₈ are independently -H, -F, -Cl, -Br, -I, -NO₂, -CN, -CO₂H, -SO₃H, -NHNH₂, -SH, -OH, -NH₂, or substituted or

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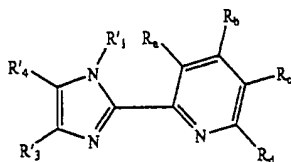
be easier or more cost-effective to make synthetically or use more widely available or more cost-effective reagents in synthesis than other transition metal redox mediators.

Compounds having Formula 1, set forth below, are examples of transition metal complexes of the present invention.



M is a transition metal and is typically iron, cobalt, ruthenium, osmium, or vanadium. Ruthenium and osmium are particularly suitable for redox mediators.

L and L' are each bidentate, substituted or unsubstituted 2-(2-pyridyl)imidazole ligands having the Structure 2 set forth below.



In Structure 2, R₁ is a substituted or an unsubstituted aryl, alkenyl, or alkyl. Generally, R₁ is a substituted or an unsubstituted C1-C12 alkyl or alkenyl, or an aryl, such as phenyl, optionally substituted with a substituent selected from a group consisting of —Cl, —F, —CN, amino, carboxy, C1-C6 alkyl, C1-C6 alkylthio, C1-C6 alkylamino, C1-C6 dialkylamino, C1-C6 alkylaminocarbonyl, C1-C6 alkoxy, C1-C6 alkoxy carbonyl, and C1-C6 alkylcarboxamido. R₁ is typically methyl or a C1-C12 alkyl that is optionally substituted with a reactive group, or an aryl optionally substituted with C1-C2 alkyl, C1-C2 alkoxy, —Cl, or —F.

Generally, R₃, R₄, R₅, R₆, R₇, and R₈ are independently —H, —F, —Cl, —Br, —I, —NO₂, —CN, —CO₂H, —SO₃H, —NHNH₂, —SH, —OH, —NH₂, substituted or unsubstituted alkoxycarbonyl, alkylaminocarbonyl, dialkylaminocarbonyl, alkoxy, alkylamino, dialkylamino, alkanoylamino, arylcarboxamido, hydrazino, alkylhydrazino, hydroxylamino, alkoxylamino, alkylthio, alkenyl, aryl, or alkyl. Alternatively, R₃ and R₄ in combination and/or R₅ and R₆ in combination can form a saturated or unsaturated 5- or 6-membered ring. Typically, the alkyl and alkoxy portions are C1 to C12. The alkyl or aryl portions of any of the substituents are optionally substituted by —F, —Cl, —Br, —I, alkylamino, dialkylamino, trialkylammonium (except on aryl portions), alkoxy, alkylthio, aryl, or a reactive group. Generally, R₃, R₄, R₅, R₆, R₇, and R₈ are independently —H or unsubstituted alkyl groups. Typically, R₃ and R₄ are —H and R₅, R₆, R₇, and R₈ are —H or methyl.

Preferably, the L and L' ligands are the same. Herein, references to L and L' may be used interchangeably.

In Formula 1, c is an integer indicating the charge of the complex. Generally, c is an integer selected from -1 to -5 or +1 to +5 indicating a positive or negative charge or 0 indicating a neutral charge. For a number of osmium complexes, c is +1, +2, or +3.

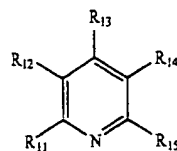
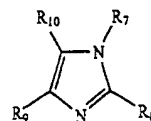
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X represents counter ion(s). Examples of suitable counter ions include anions, such as halide (e.g., fluoride, chloride, bromide or iodide), sulfate, phosphate, hexafluorophosphate, and tetrafluoroborate, and cations (preferably, monovalent cations), such as lithium, sodium, potassium, tetraalkylammonium, and ammonium. Preferably, X is a halide, such as chloride. The counter ions represented by X are not necessarily all the same.

d represents the number of counter ions and is typically from 0 to 5.

L₁ and L₂ are ligands attached to the transition metal via a coordinative bond. L₁ and L₂ are monodentate ligands, at least one of which is a negatively charged monodentate ligand. While L₁ and L₂ may be used interchangeably, L₂ is generally referred to as a negatively charged ligand merely by way of convenience. Herein, the term "negatively charged ligand" is defined as a ligand in which the coordinating atom itself is negatively charged so that on coordination to a positively charged metal, the negative charge is neutralized. For example, a halide such as chloride or fluoride meets the present definition while a pyridine ligand bearing a negatively charged sulfonate group does not because the sulfonate group does not participate in coordination. Examples of negatively charged ligands include, but are not limited to, —F, —Cl, —Br, —I, —CN, —SCN, —OH, alkoxy, alkylthio, and phenoxide. Typically, the negatively charged monodentate ligand is a halide.

Examples of other suitable monodentate ligands include, but are not limited to, H₂O, NH₃, alkylamine, dialkylamine, trialkylamine, or heterocyclic compounds. The alkyl or aryl portions of any of the ligands are optionally substituted by —F, —Cl, —Br, —I, alkylamino, dialkylamino, trialkylammonium (except on aryl portions), alkoxy, alkylthio, aryl, or a reactive group. Any alkyl portions of the monodentate ligands generally contain 1 to 12 carbons. More typically, the alkyl portions contain 1 to 6 carbons. In other embodiments, the monodentate ligands are heterocyclic compounds containing at least one nitrogen, oxygen, or sulfur atom. Examples of suitable heterocyclic monodentate ligands include imidazole, pyrazole, oxazole, thiazole, triazole, pyridine, pyrazine and derivatives thereof. Suitable heterocyclic monodentate ligands include substituted and unsubstituted imidazole and substituted and unsubstituted pyridine having the general Formulas 3 and 4, respectively, as set forth below.

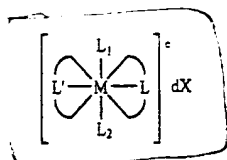


With regard to Formula 3, R₇ is generally a substituted or unsubstituted alkyl, alkenyl, or aryl group. Generally, R₇ is a substituted or unsubstituted C1 to C12 alkyl or alkenyl, or an aryl, such as phenyl, optionally substituted with a substituent selected from a group consisting of —Cl, —F, —CN, amino, carboxy, C1-C6 alkyl, C1-C6 alkylthio, C1-C6 alkylamino, C1-C6 dialkylamino, C1-C6 alkylaminocarbonyl, C1-C6 alkoxy, C1-C6 alkoxy carbonyl, and C1-C6 alkylcarboxamido. R₇ is typically methyl or a

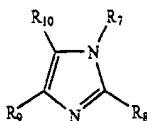
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The invention claimed is:

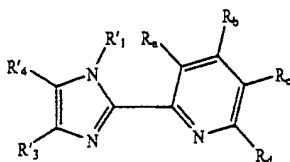
1. A complex having the formula:



wherein M is osmium;

L₁ has the formula:wherein R₇ is a substituted or unsubstituted aryl or a substituted or unsubstituted C1-C12 alkyl;R₈ is —H or a Cl alkyl; anda combination of R₉ and R₁₀ forms a fused, saturated or unsaturated, 5- or 6-membered ring;L₂ is a halide;

L and L' are independently:

wherein R'₁ is a substituted or unsubstituted C1-C6 alkyl;R'₃ and R'₄ are independently —H; andR'₂, R'₅, R'₆, and R'₇ are independently —H or Cl alkyl;

c is a +1 or +2 charge;

X is an anion; and

d is a number of X sufficient to balance the charge c.

2. The complex of claim 1, wherein R₇ is a C5 alkyl.3. The complex of any one of claims 1 and 2, wherein R₈ is methyl.4. The complex of any one of claims 1 and 2, wherein a combination of R₉ and R₁₀ forms a fused, saturated or unsaturated, 6-membered ring.5. The complex of any one of claims 1 and 2, wherein a combination of R₉ and R₁₀ forms a fused, unsaturated, 6-membered ring.6. The complex of any one of claims 1 and 2, wherein L₂ is —F, —Cl, or —Br.7. The complex of any one of claims 1 and 2, wherein L₂ is —Cl.8. The complex of any one of claims 1 and 2, wherein R'₁ is a C1-C2 alkyl.9. The complex of any one of claims 1 and 2, wherein R'₁ is a Cl alkyl.10. The complex of any one of claims 1 and 2, wherein each of R₉ and R₁₀ is —H.11. The complex of any one of claims 1 and 2, wherein each of R₉, R₁₀, and R_c is —H.

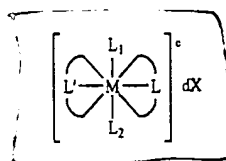
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12. The complex of any one of claims 1 and 2, wherein each of R_a, R_b, R_c, and R_d is —H.

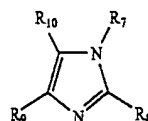
13. The complex of any one of claims 1 and 2, wherein X is a halide.

14. The complex of any one of claims 1 and 2, wherein X is chloride.

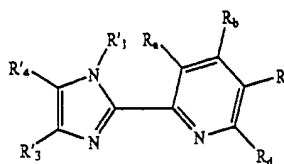
15. A complex having the formula:



wherein M is osmium;

L₁ has the formula:wherein R₇ is a C1-C 12 alkyl;R₈ is —H; anda combination of R₉ and R₁₀ forms a fused, unsaturated, 6-membered ring;L₂ is chloride;

L and L' are independently:

wherein R'₁ is methyl; andR'₃, R'₄, R'₅, R'₆, R'₇, and R'₈ are independently —H;

c is +2;

d is 2; and

X is chloride.

16. The complex of claim 15, wherein R₇ is a C5 alkyl.17. The complex of any one of claims 1, 2, 15, and 16, wherein at least one ligand selected from L, L', L₁ and L₂ is coupled to a polymeric backbone.

18. The complex of claim 17, wherein the polymeric backbone comprises at least one functional group that is a ligand of the complex.

19. The complex of claim 18, wherein the functional group is selected from pyridine and imidazole groups.

20. A sensor comprising:

a working electrode;

a counter electrode;

an enzyme disposed proximate to the working electrode; and